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AN OPTICAL STUDY OF GRAIN FORMATION: CASTING AND SOLIDIFICATION TECHNOLOGY (2-IML-1)

Mary H. McCay
University of Tennessee Space Institute
Tullahoma, TN USA

This investigation is directed at characterizing alloy solidification by studying the unidirectional growth of a metal-model material in microgravity. Using holograms and supporting temperature measurements obtained during processing in the Fluids Experiment System (FES), the solute and thermal fields associated with the dendrite growth front and extraneous nucleation will be measured and compared to a theoretical (computational) model. Ground based supporting experiments include particle tracking to measure the velocity fields, and optical phase shift techniques (e.g., confocal optical signal processing, interferometery, and Schlieren) to study thermal and solutal fields.

It is planned to utilize three cells, or CAST science modules (CSM), on Spacelab. In two CSMs a matrix of temperature gradients and growth rates will produce a variety of desired solidification conditions such as thermal undercooling, constitutional supercooling and fluid motion. In the third CSM, a dilute control (nonsolidifying) solution will be processed under two of the previous conditions. When the solidification process has been adequately characterized and compared with the model, a second series of follow-on experiments will be proposed for a future flight in which a magnified image will be used to study coarsening effects with forced fluid motion used to artificially create perturbing crystallites ahead of the interface.

The accumulation of metal model data on constitutional and thermal effects during unidirectional solidification of an alloy material is intended to enhance and then validate theoretical models of the solidification process. The microgravity data will permit the metallurgical aspects of the model to be independently verified by minimizing the overwhelming effect of convection at significant gravity levels. The validated computational models will be subsequently applied to metal systems in both environments.

A science team has been assembled that supplies expertise in the areas of microgravity solidification, fluid flow, and heat transfer, dendrite analysis, applied optics, image analysis, and the FES. Using the capabilities of this group, both the design and analysis of the experimental results will be optimized.

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Background

Constitutional Supercooling

In recent years a series of Space Processing Applications Rocket (SPAR) and KC-135 aircraft experiments have been flown to study the effects of gravity on the solidification of first, a metal-model (28 NH_4Cl-H_2O)[1,2] and then metal alloys (Sn-15Pb, Sn- 3Bi, Al-4.5Cu, MAR-M246, and PWA 1480).[3-6] In the first metal-model experiment (bi-directional solidification) four [110] dendrite arrays nucleated and grew outward from the container walls filling the entire crucible. No crystallite appeared ahead of the interface, implying that in the absence of gravity, the forces were not present that would cause dendrite fragmentation and movement in the liquid. The second (unidirectional solidification) experiment also began [110] growth at the cooled wall, but several [100] crystallites appeared and grew ahead of the interface. In each instance the low-gravity arrays grew significantly slower than one-g ground runs. In the two metal-model experiments, the growth rates were the same, but the temperature gradients differed by a factor of 7 (70 °C/cm for flight one and 10 °C/cm for flight two). This gives constitutionally supercooled regions of 0.5 cm and 3.8 cm, respectively. Based on theories of the columar to equiaxed transition in castings,[7-9] several interpretations can be made from these results, the principal two being that gravity driven fluid flow is responsible for the melting off of dendrite arms and their transport into the melt, or a non-convective related phenomena encourages nucleation ahead of the interface. Rather than distinguishing between the two possibilities as was intended, the SPAR experiments were inconclusive.

A third explanation was the occurrence of residual flow from the de-spin of the sounding rocket. If these were still present when the second experiment began freezing, they could account for crystallites moving into the fluid. This hypothesis was tested in KC-135 and F104 low gravity aircraft experiments[10,11] using shadowgraph, interferometry, and Schlieren techniques. These optical techniques enabled the experimenters to observe both the diffusion layer adjacent to the growth interface and the presence of convection plumes. The flight profile of the KC-135 causes a sample to experience 1 to 2 g's before entering a 20 sec period of low gravity. During unidirectional solidification studies growth plumes were established during the initial high g and flow rates on the order of a 1 cm/min were seen prior to inception of the low (.01 g) gravity period. These plumes dampened and began to diffuse 10 sec after entry into low gravity, indicating that the predicted[12] damping times for these experimental cases must have been inaccurate. Further thermal data from F104 flights support the concept that damping times in these materials are rapid. It, therefore, is implausible that the nuclei were carried ahead of the interface by residual flows.

Experiments with metal systems on SPAR produced similar results.[3,4] The Sn-15 Pb alloy solidified with large equiaxed grains in contrast to the more columnar plus small equiaxed grains obtained from 1-g and centrifuge solidification. The Sn-3Bi alloy also solidified initially with large grains on SPAR but the final region surrounding the shrinkage cavity froze with small equiaxed grains. It is not known if these small grains formed due to constitutional supercooling nucleation, a gravity independent flow as a result of the shrinkage cavity, or gravity driven convection due to the sounding rocket leaving the low-gravity conditions. The controversy remains concerning the formation of grains ahead of an interface.

These investigators feel that this controversy can be resolved by a more thorough study of solidification in microgravity. The extended times available on Spacelab will allow experiment conditions that cover a range of growth rates, temperature gradients, and hence, constitutionally supercooled regions without the confusing presence of gravity driven fluid flow. The solutal and thermal fields associated with the dendrite growth front will be measured and compared to a theoretical (computational) model.

Freckling

The ammonium cloride-water system is used to study the occurrence of thermalsolutal convection and pluming as it would occur in metal systems such as steel castings,[13,14] the superalloys,[15] or others[16,17] since it has an inverted diffusion layer at the interface due to rejection of water-rich solute. The pluming phenomena which occurs when the inverted layer becomes unstable is often called freckling and tends to limit the range of compositions for many alloys processed on Earth due to the resultant localized segregation and small equiaxed grains in the final casting. The plumes contain cooler liquid with a different composition from the surrounding region. As they traverse the dendrite forest, they carry dendrite fragments which appear as trails of equiaxed grains in the final ingot. The region of the freckles, therefore, has a different composition (and melting point) and crystalline morphology from the remainder of the ingot.

Freckling was first studied systematically in the metal-model NH_4C1-H_2O[15] and found to depend on thermal diffusivity, solid/liquid density ratio, solid solute solubility, solute diffusivity, and viscosity. Several investigators[15,16] have attempted to define criteria for the convective instabilities produced by the inverted layer based on either thermal properties or concentration effects. Since such criteria do not include both the effect of latent heat and segregation simultaneously, they grossly underestimate the solidification conditions (growth rate and temperature gradient) that are necessary to eliminate freckling. The presence of latent heat increases the size of the inverted layer at a critical growth rate and consideration of the combined effects decreases the growth rate for stability in NH_4C1-H_2O by 2 orders of magnitude.[18] Freckling has generally been thought to begin within the mushy zone below the dendrite tips but recent work[17] confirmed by the author also on NH_4Cl-H_2O, suggests that the channels for freckling originate at the dendrite front and spread.

Using confocal laser optical signal processing and neutrally buoyant particle laser tracking techniques, the CAST investigators studied the formation and breakdown of the inverted density layer at the dendrite front.[19,20] The convective pluming was shown not to be a natural occurrence resulting from a fundamental (Rayleigh-Benard) fluid dynamic instability at the density inversion interface. The nature of the breakdown was vortical, bounded by the dendritic interface on the bottom and the thermally lightened fluid on top (Figure 1). The significant variation with previous thinking, however, was the observation of a vortical front which serves to replenish solute without significant disturbance to the bulk liquid.

In the proposed flight experiment, the size and characteristics of the inverted layer as a function of growth condition will be measured from reconstructed holograms and compared with theoretical expectations. For the minimum experimental temperature gradient (2 °C/cm) the layer will decrease with increasing growth rate (R) until a critical R is reached upon which the layer will increase due to latent heat effects. For the maximum temperature gradient (28 °C/cm), the layer will decrease monotonically with increasing R since the critical R cannot be reached due to FES limitations.

Since optical techniques such as Schlieren and interferometry easily delineate organized convective motions, the FES system is a powerful tool for studying this phenomena. The size of a stable inverted density layer can be as large as 1 cm under the proposed experiment's low temperature gradient and growth condition. In ground based experiments the layer becomes unstable long before it reaches its maximum size with the result that events (such as equiaxed growth) within the layer are difficult to study. During the IML flight the layer can reach a large enough size that such characteristics can be resolved.

Dendrite Coarsening and Tip Concentration

Dendrite coarsening is a phenomena that occurs on the micro-scale and is primarily responsible for final dendrite arm spacings by causing the dissolution and shrinkage of smaller arms and the growth of larger arms. It is a function of local solidification time and the gradients of temperature and concentration.

The SPAR experiments[1-4] and KC-135 flights[5-6] have shown a gravity-related coarsening effect on the secondary dendrite arms. Each alloy system showed greater arm spacings for the low-gravity solidification. In the instance of KC-135 flights, the arm spacings increased in low-g, decreased in high-g, and then increased again when the next low-g parabola was flown. Theories on dendrite structure[21,22] suggest that by changing the surrounding concentration field and effective diffusion length, the perturbation frequency and, hence, the dendrite arm spacing is affected. In the case of low gravity, the diffusion length and therefore the arm spacings would increase. A more recent KC-135 experiment[5] on the superalloy PWA 1480 has shown the same physical results for the primary arm spacings (e.g., spacing increases as gravity level decreases). Current theories[23-25] suggest that these spacings are related to temperature gradient and growth rate or concentration gradient. This was the first study of low-gravity primary arm spacings, and it suggests that data at significantly increased low-gravity time periods are needed.

The present resolution of FES precludes secondary arm spacing measurements until late in the solidification runs. Hence it is planned that first the solidification process itself will be modeled for the microgravity environment, then the coarsening of the dendrite arms will be studied. The analysis of coarsening will be accomplished using a magnification lens attachment to the FES in an experiment is proposed for a subsequent flight.

Method and Approach

A two component system is required in order to model alloy solidification and investigate freckling phenomena. Since NH_4Cl-H_2O has been extensively used for similar studies, it has been chosen for these experiments. The present investigator has characterized NH_4Cl-H_2O optically in addition to accummulating other significant property data. Compared to other available metal models (e.g., succinonitrile with solute), NH_4Cl-H_2O is the superior medium available.

The CAST experiment will proceed systematically with a matrix of nine temperature gradients and growth rate conditions that will encompass a range of inverted density layer sizes and constitutionally supercooled region sizes. Based on earlier KC-135 results, the temperature gradients are expected to be different in low gravity for the same test parameters, so two non-solidifying control samples will be run at identical conditions to two of the primary runs to evaluate those differences. To produce more rapid solidification fronts, two runs will be processed in which the fluid is cooled below its freezing point and solidification initiated by thermal shock.

Holograms will be taken at repeated intervals during the growth runs. In this way, various optical techniques can be used through post-flight reconstructions to determine concentration and thermal profiles, observe perturbations in the inverted layer, and distinguish nuclei that form ahead of the interface. The techniques for reconstruction and optical analysis have been developed in support of the holographic measurements.[26-27]

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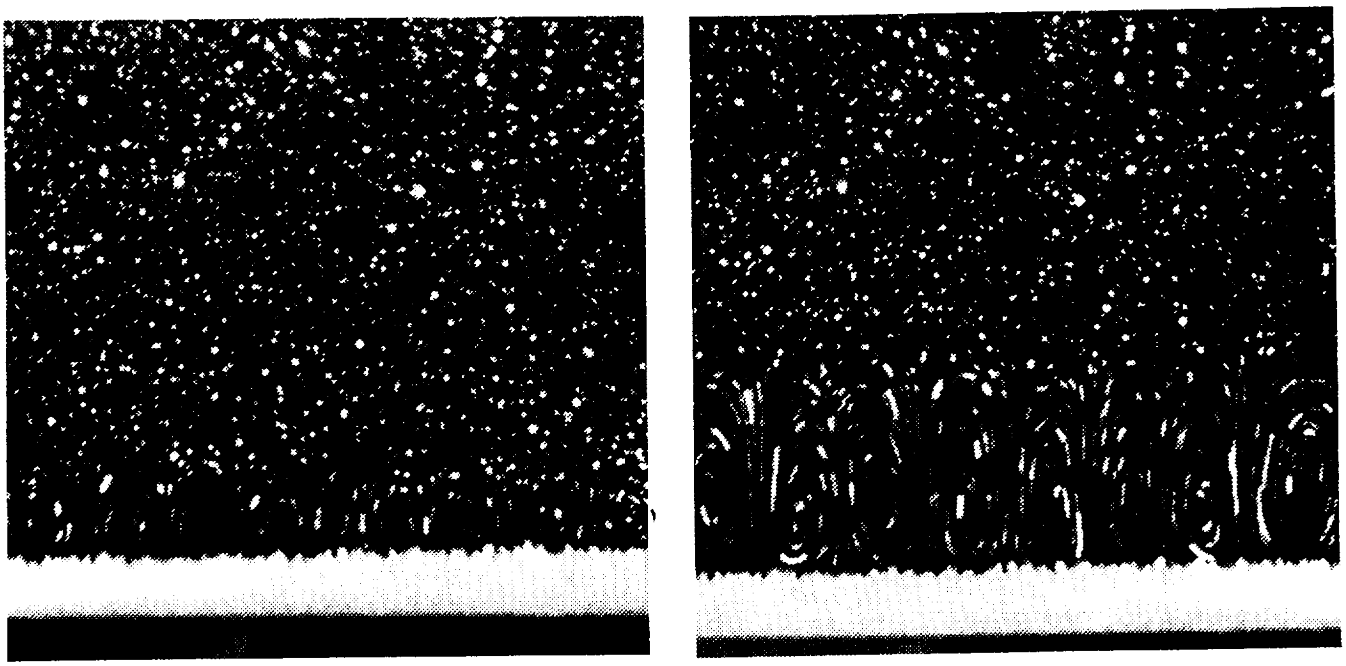

(a) (b)

Figure 1. Particle tracking optical system image of the flow geometry of a directionally solidified NH_4Cl-H_2O solution at (a) breakdown and (b) 30 seconds later.